

s –wave symmetry along the c –axis and $s + d$ in-plane superconductivity in bulk $\text{YBa}_2\text{Cu}_4\text{O}_8$

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Abstract. - To clarify the order parameter symmetry of cuprates, the magnetic penetration depth λ was measured along the crystallographic directions a , b , and c in single crystals of $\text{YBa}_2\text{Cu}_4\text{O}_8$ via muon spin rotation. *This method is direct, bulk sensitive, and unambiguous.* The temperature dependences of λ_a^{-2} and λ_b^{-2} exhibit an inflection point at low temperatures as is typical for two-gap superconductivity (TGS) with $s + d$ –wave character in the planes. Perpendicular to the planes a pure s -wave gap is observed thereby highlighting the important role of c -axis effects. We conclude that these are generic and universal features in the bulk of cuprates.

Two gap superconductivity (TGS) remained a theoretical issue only for more than 20 years [1–3], even though it seemed to be a natural and intriguing extension of BCS theory for more complex materials. In 1980 TGS was finally observed in Nb doped SrTiO_3 [4] and believed to be a rare exception in superconductors. Since the discovery of TGS in MgB_2 many more superconductors with TGS were found, including heavy fermion compounds, making this feature more common than believed early on. In all above mentioned superconductors the combined order parameters exhibit always the same symmetry, namely $s + s$ in Nb doped SrTiO_3 [4] and MgB_2 [5] and $d + d$ in heavy fermion compounds [6]. A totally novel situation is met in cuprate high-temperature superconductors (HTS), since the order parameters are of different symmetries, i.e., $s + d$ [7–13]. However, theoretical modelling suggested a single d –wave order parameter in the CuO_2 planes and unfortunately biased further research and partly inhibited the experimental efforts to characterize TGS in more detail in HTS.

Recently, new muon spin rotation (μSR) investigations of single crystal $\text{La}_{1.83}\text{Sr}_{0.17}\text{CuO}_4$ detected an inflection point in the temperature dependence of the inverse-squared in-plane magnetic penetration depth λ_{ab}^{-2} , which is a direct consequence of two superconducting gaps with

largely different zero temperature gap values [14]. Since it is unclear whether these observations are a material specific property or generic and intrinsic to HTS, the previous μSR studies were extended to single crystal $\text{YBa}_2\text{Cu}_4\text{O}_8$ and were performed by applying a magnetic field along the crystallographic directions a , b , and c . Thereby we obtain the three principle components of the second moments of the local magnetic field distribution $P(B)$ in the mixed state, which are related to the superfluid density, and reflect directly the corresponding penetration depths λ_a , λ_b , and λ_c . While λ_a^{-2} and λ_b^{-2} vary almost linearly with temperature for $20 < T < 50$ K, as is expected for a d –wave order parameter, the c –axis response (λ_c^{-2}) saturates below 30 K, as expected for a s –wave order parameter. In addition, λ_a^{-2} and λ_b^{-2} , both exhibit an inflection point in their temperature dependences around $T \simeq 10$ K which – as has been shown in [15] – is the consequence of two coexisting order parameters, namely $s + d$.

Details of the sample preparation for $\text{YBa}_2\text{Cu}_4\text{O}_8$ can be found elsewhere [16]. All crystals used in the present study were taken from one batch. The superconducting transition temperature T_c and the width of the superconducting transition ΔT_c were determined for the three sets ($\simeq 40$ -50 crystals each) of the main set ($\simeq 130$) of crys-

tals. Both were obtained from field-cooled (0.5 mT) magnetization curves measured by a SQUID magnetometer and exhibited all the same values, i.e., $T_c \simeq 79.9$ K and $\Delta T_c \simeq 2$ K. The crystals had mostly a rectangular shape with a typical size of approximately $0.8 \times 0.3 \times 0.05$ mm³. X-ray measurements revealed that the crystallographic b -axis is exactly parallel to the longest side. Bearing in mind that the c -axis is perpendicular to the flat surface of the crystal, we were able to orient the whole set along the crystallographic a , b , and c directions. The final orientation of the crystals in the mosaic was checked by using a polarizing microscope.

The transverse-field μ SR experiments on a mosaic of oriented YBa₂Cu₄O₈ single crystals were done at the π M3 and π E1 beam lines at the Paul Scherrer Institute (Villigen, Switzerland). The mosaic was field cooled from above T_c to 1.7 K in a field of 0.015 T. The μ SR experiments were performed for the magnetic field applied parallel to the a , b , and c crystallographic axes. Typical counting statistics were ~ 24 -30 million muon detections over three detectors. In the analysis presented below we used the well-known fact that for an extreme type-II superconductor in the mixed state λ^{-4} is proportional to the second moment of $P(B)$ probed by μ SR [17]. The second moment of $P(B)$ was calculated within the same framework described in [14]. We used a four component Gaussian expression to fit the μ SR time spectra. One component arises here from the background signal stemming from muons stopped outside the sample, whereas the other three components describe the asymmetric line shape of $P(B)$ in the mixed state (see inset to Fig. 1). The first and the second moments of $P(B)$ (excluding the background component) are then obtained as:

$$\langle B \rangle = \sum_{i=1}^3 \frac{A_i B_i}{A_1 + A_2 + A_3} \quad (1)$$

and

$$\langle \Delta B^2 \rangle = \frac{\sigma^2}{\gamma_\mu^2} = \sum_{i=1}^3 \frac{A_i}{A_1 + A_2 + A_3} \left[\frac{\sigma_i^2}{\gamma_\mu^2} + [B_i - \langle B \rangle]^2 \right]. \quad (2)$$

Here $\gamma_\mu = 2\pi \times 135.5342$ MHz/T is the muon gyromagnetic ratio. A_i , σ_i , and B_i are the asymmetry, the relaxation rate, and the first moment of the i -th component, respectively. The superconducting part of the square root of the second moment ($\sigma_{sc} \propto \lambda^{-2}$) was then obtained by subtracting the nuclear moment contribution (σ_{nm}) measured at $T > T_c$, according to $\sigma_{sc}^2 = \sigma^2 - \sigma_{nm}^2$. To ensure that the increase of the second moment of the measured μ SR signal below T_c is attributed entirely to the vortex lattice, zero-field μ SR experiments were performed. No evidence for static magnetism in the YBa₂Cu₄O₈ mosaic sample down to 1.7 K was observed.

For an anisotropic London superconductor the effective penetration depth for the magnetic field along the i -th

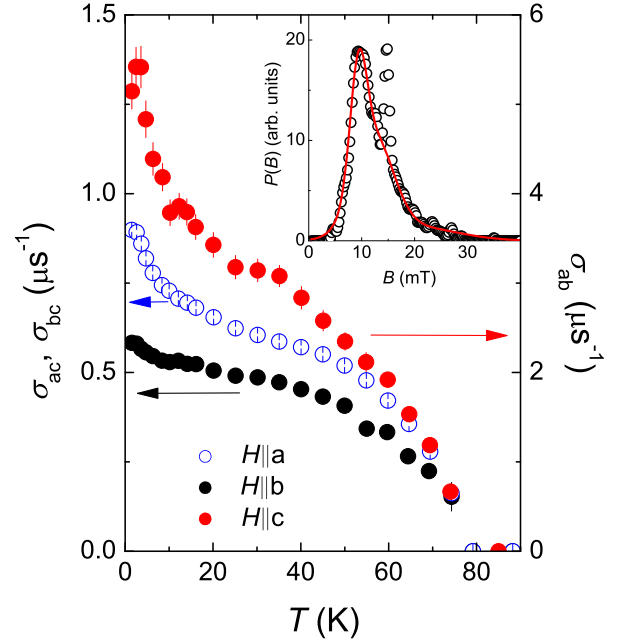


Fig. 1: (Color online) Temperature dependences of $\sigma_{ab} \propto \lambda_{ab}^{-2}$ ($H \parallel c$), $\sigma_{ac} \propto \lambda_{ac}^{-2}$ ($H \parallel b$), and $\sigma_{bc} \propto \lambda_{bc}^{-2}$ ($H \parallel a$) of YBa₂Cu₄O₈, measured after field cooling the sample in $\mu_0 H = 0.015$ T. The inset shows the local magnetic field distribution $P(B)$ obtained by means of the maximum entropy Fourier transform technique at $T = 1.7$ K and $\mu_0 H = 0.015$ T applied parallel to the c -axis (open circles: data; solid line: four component Gaussian fit excluding the background).

crystallographic axis is given by [18]:

$$\lambda_{jk}^{-2} = \frac{1}{\lambda_j \lambda_k} \propto \sigma_{jk}. \quad (3)$$

Here the index "sc" for the superconducting part of the square root of the second moment σ_{sc} is omitted for simplicity. From Eq. (3) it is obvious that for the magnetic field applied along one of the principal axes a , b , and c , the components $\sigma_{bc} \propto \lambda_{bc}^{-2}$, $\sigma_{ac} \propto \lambda_{ac}^{-2}$, and $\sigma_{ab} \propto \lambda_{ab}^{-2}$ are measured. The temperature dependences of σ_{bc} , σ_{ac} , and σ_{ab} after field-cooling the sample in $\mu_0 H = 0.015$ T are shown in Fig. 1. It is seen that at $T_{ip} \sim 10 - 20$ K all measured $\sigma_{ij}(T)$ exhibit an inflection point. Below this point σ_{ab} , σ_{bc} , and σ_{ac} increase by approximately 70%, 35%, and 10%, respectively. Note, that a similar inflection point in $\lambda_{ab}^{-2}(T)$ was also observed in low-field magnetization (LFM) experiments on powder samples of YBa₂Cu₄O₈ [19]. However, in these experiments the increase of λ_{ab}^{-2} below T_{ip} was much less pronounced than the one observed in the present study. This difference can be explained by the fact that LFM probes the penetration depth mainly near the surface, whereas μ SR measures λ in the *bulk*. In LFM experiments the magnetic field penetrates the sample only on a distance λ from the surface of the sample (few hundred nanometers), thereby leaving the main part of the superconducting volume unaffected. In contrast, muons penetrate at much longer distances into

the sample (few-tenth of a millimeter), thus probing λ deeply inside the sample. It should be recalled that at the surface due to the symmetry breaking, the order parameter has pure d -wave character, whereas in the bulk $s + d$ -wave symmetry is present [11]. This explains in a natural way the difference between the surface sensitive LFM and the bulk sensitive μ SR experiments.

From Eq. (3) the individual components of λ along the i -th crystallographic direction are obtained as:

$$\sigma_i = \frac{\sigma_{ij}\sigma_{ik}}{\sigma_{jk}} \propto \frac{1}{\lambda_i^2}. \quad (4)$$

Figure 2 shows the temperature dependences of $\sigma_a \propto \lambda_a^{-2}$, $\sigma_b \propto \lambda_b^{-2}$, and $\sigma_c \propto \lambda_c^{-2}$ as derived from the data presented in Fig. 1 by using Eq. (4). In the following we discuss the temperature dependence of each particular component separately.

From Figs. 2 (a) and (b) it is evident that both $\sigma_a \propto \lambda_a^{-2}$ and $\sigma_b \propto \lambda_b^{-2}$ increase almost linearly with decreasing temperature in the range $50 < T < 20$ K. Around $T_{ip} \simeq 10$ K an inflection point is visible in the temperature dependence. Below this point an unusual increase in both quantities appears: σ_a increases by almost 20% (from $2.95 \mu\text{s}^{-1}$ at $T = 10$ K to $3.5 \mu\text{s}^{-1}$ at $T = 2.6$ K) and σ_b by 60% (from $5.2 \mu\text{s}^{-1}$ at $T = 10$ K to $8.3 \mu\text{s}^{-1}$ at $T = 2.6$ K). As has been shown in Ref. [15], an inflection point in $\lambda^{-2}(T)$ suggests the presence of at least two superconducting gaps in $\text{YBa}_2\text{Cu}_4\text{O}_8$ with very different gap values, i.e., a large gap and a small one. The same behavior was observed recently in $\text{La}_{1.83}\text{Sr}_{0.17}\text{CuO}_4$ by μ SR [14], as well as in other HTS using various techniques [9–13], supporting a two-gap behavior with the larger gap being of d -wave and the smaller one of s -wave symmetry. The above data were analyzed by assuming that an s -wave and a d -wave gap contribute to σ according to:

$$\sigma(T) = \sigma^d(T) + \sigma^s(T), \quad (5)$$

where both components are expressed like [14]:

$$\frac{\sigma(T, \Delta_0)}{\sigma(0)} = 1 + \frac{1}{\pi} \int_0^{2\pi} \int_{\Delta(T, \varphi)}^{\infty} \left(\frac{\partial f}{\partial E} \right) \frac{E dE d\varphi}{\sqrt{E^2 - \Delta(T, \varphi)^2}}. \quad (6)$$

Here, $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function, Δ_0 is the maximum value of the gap, and $\Delta(T, \varphi) = \Delta_0 \tilde{\Delta}(T/T_c) g(\varphi)$. For the normalized gap $\tilde{\Delta}(T/T_c)$ tabulated values of Ref. [21] were used. The function $g(\varphi)$ describes the angular dependence of the gap and is given by $g^d(\varphi) = |\cos(2\varphi)|$ for the d -wave gap [12] and $g^s(\varphi) = 1$ for the s -wave gap [see insets in Figs. 2 (a) and (b)]. From this analysis we obtain $\Delta_{0,a}^d = 19.41(9)$ meV, $\sigma_a^d = 3.03(2) \mu\text{s}^{-1}$, $\Delta_{0,a}^s = 0.96(5)$ meV, $\sigma_a^s = 0.48(2) \mu\text{s}^{-1}$ and $\Delta_{0,b}^d = 21.17(8)$ meV, $\sigma_b^d = 4.66(2) \mu\text{s}^{-1}$, $\Delta_{0,b}^s = 1.10(2)$ meV, $\sigma_b^s = 3.63(2) \mu\text{s}^{-1}$ for $\sigma_a(T)$ and $\sigma_b(T)$, respectively. The main results are summarized in Table 1.

The temperature dependence of $\sigma_c \propto \lambda_c^{-2}$ differs significantly from the one of the in-plane components since

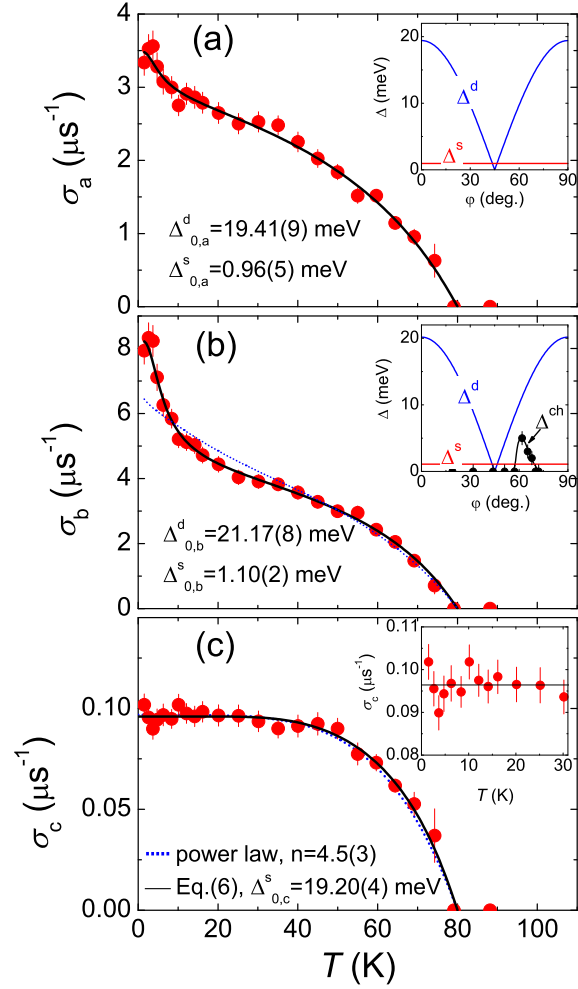


Fig. 2: (Color online) Temperature dependences of $\sigma_a \propto \lambda_a^{-2}$ (a), $\sigma_b \propto \lambda_b^{-2}$ (b), and $\sigma_c \propto \lambda_c^{-2}$ (c) of $\text{YBa}_2\text{Cu}_4\text{O}_8$ at $\mu_0 H = 0.015$ T obtained from the data presented in Fig. 1 by means of Eq. (4). Lines in (a) and (b) represent results of the fits of $\sigma_a(T)$ and $\sigma_b(T)$ by means of the two-component [Eq. (5)] and the three-component models. The blue dotted curve in (c) represents a fit to the data assuming that $\sigma_b(T)$ is entirely determined by the in-plane d -wave and the chain gap measured in [20] (see text for details). The black solid line and the blue dotted curve in (c) represent fits by means of Eq. (6) with $g^s(\varphi) = 1$ and the power law $\sigma_c(T) = \sigma_c(0)[1 - (T/T_c)^n]$, respectively. The insets in (a) and (b) represent the angular dependences of the d -wave gap (Δ^d), the s -wave gap (Δ^s) and the gap in CuO chains (Δ^{ch}) obtained in [20]. The inset in (c) shows $\sigma_c(T)$ at $T < 30$ K.

it saturates at temperatures $T < 30$ K to become T independent [see inset in Fig. 2 (c)]. This dependence is analyzed in two ways: (i) by using Eq. (6) with $g^s(\varphi) = 1$, yielding $\Delta_{0,c}^s = 19.20(4)$ meV, and (ii) by assuming the phenomenological power law $\sigma_c(T) = \sigma_c(0)[1 - (T/T_c)^n]$. The results are given in Table 1 and compared to the experimental data in Fig. 2 (c), from which it is obvious that both approaches are almost undistinguishable. For the power law dependence a critical exponent $n = 4.5(3)$

Table 1: Summary of the gap analysis of the temperature dependences of $\sigma_i \propto \lambda_i^{-2}$ ($i = a, b, c$) for $\text{YBa}_2\text{Cu}_4\text{O}_8$. The meaning of the parameters is explained in the text.

Component	$\Delta_{0,i}^d$ (meV)	$\Delta_{0,i}^s$ (meV)	$\gamma_{ab}^{\Delta^d}$	$\gamma_{ab}^{\Delta^s}$
σ_a	19.41(9)	0.96(5)	—	—
σ_b	21.17(8)	1.10(2)	1.09(1)	1.15(6)
σ_c	—	19.20(4)	—	—

is derived which is close to the one obtained within a two-fluid model where $n = 4$, which applies to a strong coupling s -wave BCS superconductors [22]. Using a d -wave model for a clean superconductor, Ref. [23] predicts an exponent $n = 5$. Since the observed power law exponent lies between these two values, it could be argued that d -wave superconductivity could also be the cause of the temperature dependence of $\sigma_c \propto \lambda_c^{-2}$. However, this can be ruled out, since the results of Ref. [23] do not apply to cuprate superconductors containing chains, like $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{YBa}_2\text{Cu}_4\text{O}_8$. We can thus safely conclude that the order parameter along the c -axis is isotropic s -wave. This conclusion is further supported by c -axis tunneling [24], bicrystal twist Josephson junctions [25], optical pulsed probe [26], and optical reflectivity (see Fig. 4 in Ref. [27]) experiments, as well as theoretical considerations [28]. It is important to note that due to the reasons described above the saturation of $\lambda_c^{-2}(T)$ obtained here has not been observed in LFM experiments [19].

The anisotropy observed in σ_a and σ_b and their unconventional temperature dependences deserve further remarks. The temperature dependences of both quantities are characterized by an inflection point at $T_{ip} \simeq 10$ K evidencing that at least two superconducting gaps contribute to the superfluid density. However, along the crystallographic b -direction this is much more pronounced than along a , and the analysis of both data sets in terms of $s + d$ wave gaps using Eq. (5) reveals that along the a direction the s -wave order parameter contributes 14%, whereas along the b direction the s -wave contribution is 42%. This observation is a consequence of the structural anisotropy caused by the CuO chains constitutes itself by an enhanced s -wave superconducting density along the chain direction which can have various origins. Early on, Raman experiments have detected the opening of a second gap at low frequencies which appears only in specific scattering geometries [29]. Since these observations have only been made in the YBCO family, they were interpreted as being due to the opening of a gap in the chains. In addition, recent angle resolved photoemission data (ARPES) have observed a coherence peak related to the chain bands, which appears in a very limited k -space region [20]. It was found that in $\text{YBa}_2\text{Cu}_4\text{O}_8$ the superconducting gap in the chains opens only at certain angles φ and reaches its maximum value of 5(1) meV at $\varphi \simeq 60^\circ$ [see inset in

Fig. 2 (b)]. Therefore, one may speculate that the two-gap behavior in $\text{YBa}_2\text{Cu}_4\text{O}_8$ is entirely determined by the in-plane d -wave and the chain gap. This scenario can, however, be excluded. First of all, the fit of Eq. (5) to $\sigma_b(T)$ taking into account a chain gap of this symmetry, leads to a rather poor agreement with the experimental data [blue dotted curve in Fig. 2 (b)]. Second, the observation of an inflection point in $\sigma_a(T)$ cannot be explained by a possible misalignment of the crystals in the mosaic. $\sigma_a^s(0) = 0.48 \mu\text{s}^{-1}$ would correspond to a situation when *all* the crystals in the mosaic are turned by $\pm 8^\circ$, or $\simeq 30^\circ$ FWHM if one assumes a triangular distribution of orientations with the maximum along the crystallographic b axis. Therefore, the data presented in Fig. 2 (b) were analyzed by assuming that $\sigma_b(T)$ is the sum of three components: $\sigma_b^d(T)$, $\sigma_b^s(T)$, and the contribution from the chains $\sigma_b^{ch}(T)$. These results are, however, undistinguishable from the $s + d$ analysis presented above. It is important to note, that independent of a possible chain related energy gap, both procedures provide clear evidence that the in-plane superconducting order parameter consists of two components, a d -wave order parameter and an additional s -wave component.

The maximum values of the in-plane d -wave gap along the a and b directions are of similar order of magnitudes, i.e., $\Delta_{0,a}^d = 19.41(9)$ meV, $\Delta_{0,b}^d = 21.17(8)$ meV and thus in a good agreement with $\Delta_0^d = 22$ meV derived from tunnelling experiment [30]. The in-plane anisotropy in both gap values, s and d , is approximately the same, i.e., $\gamma_{ab}^{\Delta^d} = \Delta_{0,b}^d/\Delta_{0,a}^d = 1.09(1)$ and $\gamma_{ab}^{\Delta^s} = \Delta_{0,b}^s/\Delta_{0,a}^s = 1.15(6)$ (see Table 1). Moreover, it is important to note that the s -wave gap along the c direction $\Delta_{0,c}^s = 19.20(4)$ meV is of the same order of magnitude as the d -wave gaps in the ab plane: $\Delta_{0,c}^s \simeq \Delta_{0,a}^d \simeq \Delta_{0,b}^d \approx 20$ meV.

In conclusion, we performed a systematic μSR study of the magnetic penetration depths λ_a , λ_b , and λ_c on single crystals of $\text{YBa}_2\text{Cu}_4\text{O}_8$. In contrast to previous LFM experiments [19], our method is *bulk* sensitive and a direct probe of the penetration depth. The use of single crystals enables us to derive the magnetic penetration depth along the three principal crystallographic directions. Along the a and b directions clear evidence is obtained that TGS is realized also in the chain containing compound $\text{YBa}_2\text{Cu}_4\text{O}_8$. While the in-plane penetration depth is anisotropic, exhibiting an inflection point at low temperatures in both λ_a^{-2} and λ_b^{-2} which is characteristic of TGS, the c -axis data provide clear evidence for an isotropic s -wave gap. From the data it must be concluded that the in-plane superconducting gap consists of two components, namely $s + d$. The situation along the c -axis is different and supports s -wave only. This exceptional behavior has not been predicted by any theory, since the third dimension has mostly been neglected. Since $\text{YBa}_2\text{Cu}_4\text{O}_8$ differs structurally substantially from the previously investigated system $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, where the in-plane penetration depth also shows $s + d$ wave super-

conductivity [14], the above findings cannot be attributed to specific structural features of the chain containing compound, but show that $s + d$ order parameters are generic, intrinsic and common to all HTS. The results thus exclude theoretical approaches as e.g., the "plain vanilla" mechanism which concentrates on the CuO_2 plane only. Besides of the fact that t - J or 2D Hubbard models are not capable to yield the observed s -wave component, the role of the lattice played for superconductivity has to be reconsidered since the presence of an s -wave component naturally points to its importance.

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